

# Identification of the Orbital Pairing Symmetry in UPt<sub>3</sub>

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This paper summarizes the results of a comprehensive analysis of the thermodynamic and transport data for the superconducting phases of UPt<sub>3</sub>. Calculations of the transverse sound attenuation as a function of temperature, frequency, polarization, and disorder are presented for the leading models of the superconducting order parameter. Measurements of the specific heat, thermal conductivity, and transverse sound attenuation place strong constraints on the orbital symmetry of the superconducting order parameter. We show that the superconducting A and B phases are in excellent agreement with pairing states belonging to the odd-parity E<sub>2u</sub> orbital representation.

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## I. INTRODUCTION

Unconventional superconductivity, the electronic analog of superfluidity in <sup>3</sup>He, was discovered in the heavy-fermion metals UBe<sub>13</sub> and UPt<sub>3</sub> more than a decade ago.<sup>1,2</sup> As in liquid <sup>3</sup>He the observation of multiple superconducting phases was the direct evidence for a multi-component superconducting order parameter.<sup>3–6</sup> The phases of UPt<sub>3</sub> have since become a paradigm for unconventional superconductivity. However, unlike the case of <sup>3</sup>He the identification of the orbital and spin symmetry of the order parameter has been a more difficult task. Heavy fermion metals are more complex materials in which disorder, magnetism, spin-orbit coupling and anisotropy must be factored into any realistic theory of superconductivity in these systems (c.f. Refs. 7,8).

In this paper we present new theoretical results and analysis of the transport properties of the leading models for the superconducting phases of UPt<sub>3</sub>. These models yield qualitatively different predictions for the transport properties in the superconducting phases. We calculate the ultrasonic attenuation for the A and B phases and discuss its sensitivity to order parameter symmetry, polarization direction and disorder. From our analysis of experimental data for the heat capacity,<sup>9</sup> thermal conductivity<sup>10,11</sup> and transverse sound attenuation<sup>12</sup> we determine the topology of the excitation gap on the Fermi surface and conclude that the orbital symmetry of the order parameter in the A and B phases of UPt<sub>3</sub> belongs to an odd-parity E<sub>2u</sub> representation.

## II. PAIRING SYMMETRY

The discoveries of multiple superconducting phases<sup>3–5,13,14</sup> of UPt<sub>3</sub> led to several theoretical models for the superconducting phase diagram based on different

symmetry groups, or symmetry breaking scenarios.<sup>15–19</sup> One class of models is based on a two-dimensional (“E”) representation of the hexagonal point group, D<sub>6h</sub>, with the multi-component superconducting order parameter coupled to a *symmetry breaking field* (SBF). There are four E-representations for strong spin-orbit coupling: two E-reps for both even-parity (E<sub>1g</sub>, E<sub>2g</sub>) and odd-parity (E<sub>1u</sub>, E<sub>2u</sub>) pairing. The E-rep models require a weak SBF that lowers the symmetry of the normal state, splits the superconducting transition and produces multiple superconducting phases.<sup>15</sup> The SBF is generally assumed to be the in-plane antiferromagnetic order parameter that onsets at  $T_N \simeq 5\text{ K}$ ;<sup>20</sup> however, other explanations of the SBF have been suggested.<sup>21–23</sup> The precise structure of the short-range AFM correlations, e.g. the spatial structure of domains, as well as the role of AFM as a SBF for superconductivity is still an open question.<sup>24</sup> One of the outcomes of the calculations summarized below is that simple model of equal-size, equally populated multi-domain structures for the SBF is in disagreement with the anisotropy of the sound attenuation.

The models that have been most successful in explaining the properties of the superconducting phases of UPt<sub>3</sub> are based on the even-parity (E<sub>1g</sub>) and the odd-parity (E<sub>2u</sub>) representations of the hexagonal point group. The E<sub>1g</sub> representation is a realization of spin-singlet, d-wave pairing for a metal with a uniaxial symmetry, while the E<sub>2u</sub> model describes the hexagonal analog of spin-triplet, f-wave pairing. These pairing states have an orbital order parameter of the form  $\Delta(\mathbf{p}_f) = \eta_1 \mathcal{Y}_1(\mathbf{p}_f) + \eta_2 \mathcal{Y}_2(\mathbf{p}_f)$ , where  $\mathcal{Y}_{1,2}(\mathbf{p}_f)$  are the basis functions for the appropriate E-representation, and the amplitudes  $\boldsymbol{\eta} = (\eta_1, \eta_2)$  transform as a two-component ‘vector’ under the same E-representation. Thus, in the E-representations the order parameter of the A phase is identified as  $\boldsymbol{\eta} = (1, 0)$ , the B phase as  $\boldsymbol{\eta} = (1, i)$ , and the C phase as  $\boldsymbol{\eta} = (0, 1)$  (see Fig. 1). These identifications then refer to the specific basis functions,  $\mathcal{Y}_{1,2}(\mathbf{p}_f)$ , for a particular E-representation

given in Table I. The orbital order parameter differs significantly for the two models, particularly for the high temperature A phase ( $\eta_2 = 0$ ). For  $E_{1g}$  pairing the A phase has the structure,  $\Delta_A \sim p_z p_x$ , which has an equatorial line node in the basal plane, as well as a longitudinal line node circumscribing the Fermi surface. For the  $E_{2u}$  representation,  $\Delta_A \sim p_z (p_x^2 - p_y^2)$  also has an equatorial line node, but has two longitudinal line nodes oriented 90 degrees to one another. The low-temperature B phase of both models breaks time-reversal symmetry (with  $\eta_2 \simeq \pm i\eta_1$ ). As a result the longitudinal line nodes are closed by the growth of the second component of the order parameter; and for  $T \rightarrow 0$ ,  $\Delta_B \sim p_z(p_x + ip_y)$  for  $E_{1g}$  symmetry, while  $\Delta_B \sim p_z(p_x + ip_y)^2$  for the  $E_{2u}$  representation. Thus, the low energy excitation spectra for the B phase of the  $E_{1g}$  and  $E_{2u}$  models is described by an equatorial line of zero energy excitations ( $p_z = 0$ ) and pairs of point nodes of the excitation gap ( $p_x = p_y = 0$ ) on the Fermi surface. There is a slight difference in the density of states from the point nodes because the gap varies linearly near the point nodes for the  $E_{1g}$  model,  $|\Delta(\mathbf{p}_f)| \sim |\vartheta|$ , but quadratically for the  $E_{2u}$  model,  $|\Delta(\mathbf{p}_f)| \sim |\vartheta|^2$ . These slight differences are predicted to be observable in the heat transport at ultralow temperatures.<sup>25–27</sup>

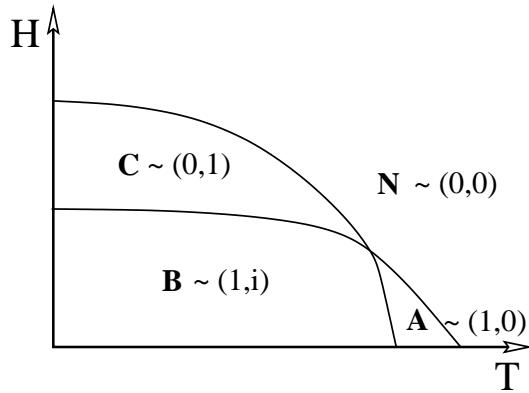


FIG. 1. The phase diagram of  $\text{UPt}_3$ . The three superconducting phases A, B, and C with amplitudes  $\eta = (\eta_1, \eta_2)$  meet with the normal state (N) at the tetracritical point. For simplicity the additional two Meissner phases are not shown.

All of the E-rep models are based on two-component orbital order parameters. However, they yield different predictions for the thermodynamic, magnetic and transport properties, including the H-T phase diagram. One important difference arises for the case of weak in-plane hexagonal anisotropy, as is reflected by the very small in-plane anisotropy of  $H_{c2}$ .<sup>28,29</sup> Weak in-plane anisotropy leads to an apparent tetracritical point for all field orientations provided the order parameter belongs to an  $E_2$  orbital representation.<sup>7</sup>

A key difference between the even-parity and odd-parity E-representations is the spin structure of the order parameter. Odd-parity representations are spin-

triplet pairing states, and in the absence of spin-orbit coupling the dimensionality of these representations is three times larger than that of the corresponding spin-singlet E-representations. However, strong spin-orbit coupling in the Uranium-based heavy fermion metals reduces the symmetry group by allowing only joint rotations of the spin and orbital degrees of freedom. The even and odd parity representations are still described by (pseudo) spin-singlet and spin-triplet order parameters of the form<sup>30–32</sup>

$$\Delta_{\alpha\beta}(\mathbf{p}_f) = \Delta(\mathbf{p}_f) (i\sigma_y)_{\alpha\beta} \quad (\text{singlet}), \quad (1)$$

$$\Delta_{\alpha\beta}(\mathbf{p}_f) = \mathbf{\Delta}(\mathbf{p}_f) \cdot (i\sigma\sigma_y)_{\alpha\beta} \quad (\text{triplet}). \quad (2)$$

The triplet representations transform only under *joint* spin and orbital rotations of the discrete point group for the normal state, i.e.,  $\mathbf{\Delta}(\mathbf{p}_f) \rightarrow \mathcal{R}\mathbf{\Delta}(\mathcal{R}^{-1}\mathbf{p}_f)$ , where the rotation  $\mathcal{R} \in [D_{6h}]_{\text{spin-orbit}}$ . The full symmetry group of the normal state is  $\mathcal{G} = [D_{6h}]_{\text{spin-orbit}} \times \mathcal{T} \times U(1)$  with  $[D_{6h}]_{\text{spin-orbit}}$  representing the hexagonal point group with inversion,  $\mathcal{T}$  is the time-inversion operation and  $U(1)$  is the group of gauge transformations. In the limit of no spin-orbit coupling  $\mathbf{\Delta}(\mathbf{p}_f)$  transforms as a spin vector under the vector representation of the full spin-rotation group, and separately as a representation of the point group with respect to the orbital momentum,  $\mathbf{p}_f$ , i.e.,  $\mathbf{\Delta}(\mathbf{p}_f) \rightarrow \mathcal{R}_{\text{spin}}\mathbf{\Delta}(\mathcal{R}_{\text{orbit}}^{-1}\mathbf{p}_f)$ , where  $\mathcal{R}_{\text{spin}} \in SU(2)_{\text{spin}}$  and  $\mathcal{R}_{\text{orbit}} \in [D_{6h}]_{\text{orbit}}$ . In the absence of spin-orbit coupling the enlarged symmetry group for the normal state is  $\mathcal{G} = SU(2)_{\text{spin}} \times [D_{6h}]_{\text{orbit}} \times \mathcal{T} \times U(1)$ .

There are two special classes of spin-triplet order parameters that are frequently discussed as candidates for the phases of  $\text{UPt}_3$ . The first class are states in which the spin-triplet order parameter factorizes into a single spin-vector and an orbital amplitude, i.e.,  $\mathbf{\Delta}(\mathbf{p}_f) = \mathbf{d}\Delta(\mathbf{p}_f)$  where  $\mathbf{d}$  is a real unit vector and  $\Delta(\mathbf{p}_f)$  is an odd-parity orbital function. The vector  $\mathbf{d}$  defines the axis along which the pairs have zero spin projection, e.g., if  $\mathbf{d} \parallel \mathbf{z}$  then  $\Delta_{\uparrow\uparrow} = \Delta_{\downarrow\downarrow} = 0$  and  $\Delta_{\uparrow\downarrow} = \Delta_{\downarrow\uparrow} = \Delta(\mathbf{p})$ . If we choose the quantization axis to be perpendicular to  $\mathbf{d}$ , i.e.,  $\mathbf{d} \perp \mathbf{z}$ , then the same pairing state is described as *equal spin pairing* in an “easy-plane”, i.e., the pairs form triplet states with amplitudes  $\Delta_{\leftarrow} = \Delta_{\rightarrow} = \Delta(\mathbf{p})$  and  $\Delta_{\leftarrow\rightarrow} = 0$ . In the second class,  $\mathbf{d}$  is complex and the spin components of the order parameter spontaneously break time-reversal symmetry. In the general case  $\mathbf{\Delta}$  is complex, with  $\mathbf{\Delta} \times \mathbf{\Delta}^* \neq 0$ , and varies over the Fermi surface. These states are called ‘non-unitary’ because the square of the spin-matrix representation of the order parameter is no longer proportional to the unit spin matrix,  $[\Delta^\dagger\Delta]_{\alpha\beta} = |\mathbf{\Delta}|^2 \delta_{\alpha\beta} + i[\mathbf{\Delta} \times \mathbf{\Delta}^* \cdot \boldsymbol{\sigma}]_{\alpha\beta}$ . As a consequence the spin degeneracy of the excitation spectrum is lifted and the quasiparticle energy depends on the local pair spin at  $\mathbf{p}_f$ :  $\mathbf{S}_{\text{pair}}(\mathbf{p}_f) \sim i\mathbf{\Delta}(\mathbf{p}_f) \times \mathbf{\Delta}(\mathbf{p}_f)^*$ .

Whether or not spin-orbit coupling is weak or strong on the energy scale of  $k_B T_c$  has important implications for both the orbital and spin components of the order parameter that are allowed by symmetry. Blount<sup>32</sup> and Volovik

and Gorkov<sup>31</sup> showed that line nodes are *not required* for odd-parity states when spin-orbit coupling is relevant. However, line nodes in the  $ab$ -plane of the Fermi surface are allowed, and required for some representations, *if* the normal-state spin-orbit interactions lock  $\mathbf{d}$  along the  $\mathbf{c}$  axis of the crystal, i.e.,  $\mathbf{d} \parallel \mathbf{c}$ . Precisely this orientation of  $\mathbf{d}$  was predicted<sup>33</sup> for  $\text{UPt}_3$  based on anisotropic paramagnetic limiting.<sup>28</sup> This effect arises from the competition between the condensation energy and the Zeeman energy. For  $\mathbf{d}$  locked along the  $\mathbf{c}$  axis of the lattice the Zeeman energy is pair-breaking for  $\mathbf{H} \parallel \mathbf{c}$ , giving rise to paramagnetic limiting. However, for  $\mathbf{H} \perp \mathbf{c}$  the Zeeman energy,  $\mathcal{F}_{\text{Zeeman}} \sim (\mathbf{d} \cdot \mathbf{H})^2$ , is minimum (vanishes); as a result there is no paramagnetic limit for this field orientation. The anisotropic paramagnetic limiting of  $H_{c2}$  is sensitive to the spin structure of the order parameter, but insensitive to the orbital pairing symmetry.<sup>33,34,7,35</sup> The odd-parity  $E_{2u}$  representation with strong spin-orbit locking of  $\mathbf{d} \parallel \mathbf{c}$  quantitatively accounts for the anisotropy of the paramagnetic limit of  $H_{c2}$  observed at low temperatures.

The spin-singlet  $E_{1g}$  model appears to be incompatible with both the tetracritical point for  $\mathbf{H} \not\parallel \mathbf{c}$  and the anisotropic paramagnetic limiting of  $H_{c2}$ . However, Park and Joynt<sup>36</sup> argue that there is enough freedom in the  $E_{1g}$  model to account for the existing experimental data on  $H_{c2}$ . Both E-rep models have recently been challenged by observations of a nearly temperature independent Knight shift for  $\mathbf{H} \parallel \mathbf{c}$ ,<sup>37</sup> which is interpreted in terms of non-unitary, spin-triplet pairing with weak, or no spin-orbit coupling.<sup>38</sup> The authors of Ref. 37 assume that the Knight shift measures the bulk spin susceptibility. If, for simplicity, we ignore the anisotropy of the normal-state susceptibility, then for a given orientation of  $\mathbf{d}$  the spin susceptibility is given by  $\chi_{ij} = \chi_N (\delta_{ij} - d_i d_j) + \chi_0 d_i d_j$ , where  $\chi_N$  is the normal state spin susceptibility and  $\chi_0(T)$  is the spin susceptibility for  $\mathbf{H} \parallel \mathbf{d}$ , which is suppressed by pair-breaking and vanishes for  $T \rightarrow 0$  in the clean limit. For strong spin-orbit coupling with  $\mathbf{d}$  locked along  $\mathbf{c}$  we expect a suppression of the Knight shift for  $T < T_c$  for fields  $\mathbf{H} \parallel \mathbf{c}$ , but no suppression for  $\mathbf{H} \perp \mathbf{c}$ .

However, in the limit of no spin-orbit coupling the Zeeman energy is minimized by *rotation* of  $\mathbf{d}$  perpendicular to the field. This implies that the Knight shift will be temperature independent and given by the normal-state shift for all field orientations. The NMR measurements of the Knight shift<sup>37</sup> appear to be in conflict with anisotropic paramagnetic limiting of  $H_{c2}$ . The paramagnetic limit observed for  $H_{c2}^{\parallel}$  is a robust, thermodynamic property of bulk single crystals of  $\text{UPt}_3$ , and a consistent interpretation of the NMR results for the Knight shift data must accommodate anisotropic paramagnetic limiting. This cannot be accomplished with a model of spin-triplet pairing without strong spin-orbit coupling.

Our analysis presented below for the heat capacity and transport measurements is independent of the interpretation of the Knight shift measurements. We show that the heat capacity, low temperature thermal conductivity and transverse sound attenuation data, in addition to the H-T phase diagram, are in quantitative agreement *only* for the odd-parity  $E_{2u}$  representation (Table I), independent of the orientation of  $\mathbf{d}$ . In order to demonstrate this fact we present calculations for other models that have been proposed to account for the phase diagram. Thus, in addition to the 2D E-representations we also examine the transport properties of the order parameter models belonging to mixed representations of the  $D_{6h}$  point group, i.e., the AB models<sup>18,19</sup> and the AE model.<sup>39</sup> These models were proposed as alternatives to the E-representations to explain the Ginzburg-Landau region of the H-T phase diagram. The most promising candidate of the AB model is the odd parity, spin-triplet model with mixed  $A_{2u} \oplus B_{1u}$  symmetry. The orbital order parameter for the A phase has the form,  $\Delta_A(\mathbf{p}_f) \sim p_z \text{Im}(p_x + ip_y)^6$ , exhibiting an equatorial line node and six longitudinal line nodes. We also analyze the transport properties of the even-parity, spin-singlet  $A_{1g} \oplus E_{1g}$  model with an A phase of the form,  $\Delta_A(\mathbf{p}_f) \sim (2p_z^2 - p_x^2 - p_y^2)$ , which has a pair of ‘tropical’ line nodes located off the equatorial plane. For a more detailed description of the order parameter for these models see Refs. 39,40.

TABLE I. Polynomial functions representing the symmetry of the low-temperature B phases of several pairing models. The first three entries are based on the symmetry group  $[D_{6h}]_{\text{spin-orbit}} \times \mathcal{T} \times U(1)$ . The third entry is representative of the class of AB models, and the last entry belongs to mixed symmetry representations resulting from the crystal-field splitting of the enlarged symmetry group,  $SO(3)_{\text{spin-orbit}} \times \mathcal{T} \times U(1)$ .

$\Gamma$	$\mathcal{Y}_{\Gamma}$	point nodes	line nodes	cross nodes
$E_{1g}$	$p_z(p_x + ip_y)$	$\vartheta = 0, \pi$	$\vartheta = \frac{\pi}{2}$	—
$E_{2u}$	$p_z(p_x + ip_y)^2$	$\vartheta = 0, \pi$	$\vartheta = \frac{\pi}{2}$	—
$A_{2u} \oplus B_{1u}$	$A p_z \text{Im}(p_x + ip_y)^6$ + $i B \text{Im}(p_x + ip_y)^3$	—	$\varphi_n = n \frac{\pi}{3},$ $n = 0, \dots, 5$	$\vartheta = 0, \pi \wedge \varphi_n$
$A_{1g} \oplus E_{1g}$	$A (2p_z^2 - p_x^2 - p_y^2)$ + $i E p_y p_z$	—	—	$\vartheta = \cos^{-1} \frac{\pm 1}{\sqrt{3}}$ $\wedge \varphi = 0, \pi$

### III. TRANSPORT THEORY

Electronic transport in the superconducting state is sensitive to the nodal structure of the order parameter,  $\Delta(\mathbf{p}_f)$ . Recent theoretical analyses<sup>41,25,26,40</sup> of low-temperature thermal conductivity data on superconducting  $\text{UPt}_3$ <sup>10,11</sup> have eliminated most of the theoretical models proposed to explain the phase diagram of  $\text{UPt}_3$ . The non-unitary, spin-triplet pairing states based on a one-dimensional ( $1D$ ) orbital representation studied so far,<sup>42–45,38</sup> as well as the two-component order parameter models obtained from nearly degenerate one-dimensional representations<sup>19</sup> ('AB models'), are unable to describe, even qualitatively, the temperature dependence and anisotropy of the thermal conductivity at low temperatures. The only pairing models which can account for the thermal conductivity data are the two-dimensional ( $2D$ ) orbital representations,  $E_{1g}$  and  $E_{2u}$ , and the  $A_{1g} \oplus E_{1g}$  (AE) model. However, the AE model predicts a large  $ab$ -plane anisotropy, which has so far not been observed.<sup>40</sup> After it was shown that a non-unitary, spin-triplet state with a  $1D$  orbital basis function was incompatible with the thermal conductivity data,<sup>40</sup> Machida et al.<sup>46</sup> modified their weak spin-orbit coupling model by adopting the  $2D$  orbital representation  $E_{2u}$ . However, the model of Ref. 46 proposes a spin structure for the order parameter which is in conflict with the observed Pauli limiting for  $\mathbf{H} \parallel \mathbf{c}$ , and it predicts a fourth superconducting phase which disagrees with the phase diagram.

Transverse ultrasound is an even more powerful probe of the order parameter and excitation spectrum than the thermal conductivity.<sup>47,12</sup> The attenuation of hydrodynamic sound is determined by the electronic viscosity tensor, which is sensitive to the relative orientation of the polarization, propagation direction and order parameter.<sup>48,49</sup> The broken symmetries of the pairing state give rise to additional anisotropy of the low-energy excitation spectrum that is specific to the pairing state; the selection rules for acoustic absorption reflect these broken symmetries.

In the hydrodynamic limit,  $\omega\tau \ll 1$  and  $q\ell \ll 1$ , where  $\ell = v_f\tau$  is the quasiparticle mean-free-path and  $\tau$  is the transport collision time, the ultrasonic attenuation is determined by components of the viscosity tensor

$$\alpha(\mathbf{q}, \epsilon, T) = (\omega^2 / \varrho c_s^3) \eta_{ij,kl}(\mathbf{q}, \omega) \hat{\epsilon}_i \hat{\mathbf{q}}_j \hat{\epsilon}_k \hat{\mathbf{q}}_l, \quad (3)$$

where  $\varrho$  is the mass density,  $c_s = \omega/q$  is the speed of the sound mode with wavevector  $\mathbf{q}$  and polarization  $\epsilon$ .<sup>50,51</sup> The hydrodynamic limit is achieved even for high-purity single-crystals of  $\text{UPt}_3$ . For the experiments reported in Ref. 12 with propagation  $\mathbf{q} \parallel \mathbf{a}$  and polarization  $\epsilon \parallel \mathbf{b}$  the sound frequency is  $\omega/2\pi \simeq 165$  MHz, the speed of sound is  $c_s \simeq 2.1$  km/s,<sup>28</sup> and the elastic mean free path is  $\ell_{ab} = v_{f,ab}\tau \approx 1.5$  km/s  $\cdot$  240 ps  $\approx 360$  nm,<sup>52</sup> yielding  $\omega\tau \approx 0.25$  and  $q\ell_{ab} \approx 0.18$  at  $T = 0$ . Similarly, for  $\mathbf{q} \parallel \mathbf{a}$  and  $\epsilon \parallel \mathbf{c}$  the reported values are  $\omega/2\pi \simeq 228$  MHz,

$c_s \simeq 1.4$  km/s, and  $\ell_c \approx \sqrt{2.7}\ell_{ab}$ , yielding  $\omega\tau \approx 0.34$  and  $q\ell_c \approx 0.61$  at  $T = 0$ . The parameters  $q\ell_c$  and  $\omega\tau$  are a factor of two smaller near  $T_c$  than they are at low temperature, since  $\tau(T_c) \simeq \tau(0)/2$ . Nevertheless, attenuation measurements for  $\epsilon \parallel \mathbf{c}$  are near the borderline of the hydrodynamic regime. Measurements above and below this cross-over regime would be desirable; both for checking the applicability of hydrodynamic results for the attenuation for  $\epsilon \parallel \mathbf{c}$ , and such measurements might exhibit to new phenomena in the collisionless regime.

The viscosity and sound attenuation are calculated from the response of the momentum stress tensor to an ionic displacement field  $\mathbf{A}(\mathbf{q}, \omega) = A(\mathbf{q}, \omega)\epsilon$ . For transverse modes ( $\mathbf{q} \cdot \epsilon = 0$ ) the stress and viscosity tensors are related in the hydrodynamic limit by<sup>53,54</sup>

$$\Pi_{ij}(\mathbf{q}, \omega) = \omega q A(\mathbf{q}, \omega) \eta_{ij,kl}(\mathbf{q}, \omega) \hat{\epsilon}_k \hat{\mathbf{q}}_l. \quad (4)$$

At low temperatures the transfer of energy and momentum between the ionic lattice and electronic excitations is dominated by the scattering of quasiparticles off impurities or defects. The theory of momentum transport by quasiparticle scattering is formulated in terms of nonequilibrium Green's functions for electronic quasiparticles coupled to the acoustic modes of the lattice. The momentum stress tensor is

$$\Pi_{ij}(\mathbf{q}, \omega) = N_f \int \frac{d\epsilon}{8\pi i} \int d\mathbf{p}_f [\mathbf{v}_f]_i [\mathbf{p}_f]_j \delta g^K(\mathbf{p}_f, \mathbf{q}; \epsilon, \omega), \quad (5)$$

where  $\mathbf{v}_f$  is the Fermi velocity,  $\mathbf{p}_f$  is the Fermi momentum,  $N_f$  the density of states at the Fermi surface, and  $\delta g^K$  is the nonequilibrium quasiparticle Green's function, integrated with respect to the quasiparticle energy,  $\xi_{\mathbf{p}} \simeq v_f(p - p_f)$ ;  $\delta g^K$  includes both the changes in the distribution of occupied states and the dynamics of the spectrum of low-energy excitations (see Appendix).

### IV. TRANSPORT EQUATIONS

We use Keldysh's formulation of the nonequilibrium response theory and calculate the transport properties and related Green's functions in the quasiclassical limit, which is easily achieved in  $\text{UPt}_3$  for excitation energies,  $(k_B T, \hbar\omega) \ll E_f \approx 1$  meV, and wavelengths long compared to the Fermi wavelength, i.e.,  $\hbar q \ll p_f$ . The central equation for the nonequilibrium Green's function in the quasiclassical limit is a transport equation. For small deviations from equilibrium the transport equation may be linearized in the deviations of the Green's function from its local equilibrium form. Our analysis and notation follows that of Ref. 41, which provides a detailed discussion of the quasiclassical linear response theory, including a complete solution to the linearized nonequilibrium transport equations. A summary of these equations, applicable to momentum transport in Fermi-liquid superconductors, is given in the Appendix.

The transport equation for the Green's function includes the acceleration of electronic quasiparticles by the acoustic field and collision terms which transfer momentum between the lattice and the electrons. The stress tensor, and therefore the electronic viscosity which damps the acoustic wave, is calculated from the solution of the transport equation for the nonequilibrium Green's function,  $\delta g^K$ , which is driven by the coupling of quasiparticles to the ionic displacement field, i.e., an externally imposed sound field,  $\hat{\sigma}_{\text{ext}}(\mathbf{p}_f, \mathbf{q}; \omega) = i(\mathbf{v}_f \cdot \mathbf{q})(\mathbf{p}_f \cdot \mathbf{A})\hat{1}$ .<sup>53,54</sup> Below we report new results for the electronic shear viscosity for the order parameter models of UPt<sub>3</sub>, and new calculations of the anisotropy and temperature dependence of the attenuation which we use to interpret the experimental data for UPt<sub>3</sub>.

In the limit of  $\omega \rightarrow 0$  and for resonant scattering the viscosity tensor simplifies to

$$\eta_{ij,ij} = -\frac{N_f}{8\pi^3 k_B T} \int d\epsilon \operatorname{sech}^2(\epsilon/2k_B T) \times \int d\mathbf{p}_f \frac{[\mathbf{v}_f]_i^2 [\mathbf{p}_f]_j^2}{\operatorname{Re} C^R} [g_0^R g_0^{R*} - f_0^R f_0^{R*} + \pi^2], \quad (6)$$

where  $C^R = -\frac{1}{\pi} \sqrt{|\Delta(\mathbf{p}_f)|^2 - (\tilde{\epsilon}^R)^2}$ ,  $g_0^R = \tilde{\epsilon}^R/C^R$ ,  $f_0^R = -\Delta(\mathbf{p}_f)/C^R$ , and  $\tilde{\epsilon}^R = \epsilon - \frac{1}{4} \operatorname{Tr}[\hat{\tau}_3 \hat{\sigma}_{\text{imp}}^R]$  is the impurity-renormalized energy. In the case of triplet pairing with a unitary order parameter the only change in Eq. (6) is replacement of  $\Delta \rightarrow \Delta$  and  $f_0^R f_0^{R*} \rightarrow \mathbf{f}_0^R \cdot \mathbf{f}_0^{R*}$ . See the Appendix for details on the notation.

For a normal metal with a spherical Fermi surface we obtain Pippard's result for the viscosity,  $\eta_{ij,ij} = \frac{2}{15} v_f^2 p_f^2 N_f \tau$  for  $i \neq j$ .<sup>55</sup> Below  $T_c$  the sound attenuation drops; for a conventional superconductor, in the limit  $q\ell \ll 1$ , the attenuation decreases exponentially for  $k_B T \ll \Delta$ .<sup>53</sup> But for an unconventional superconductor in which the order parameter vanishes at points or lines on the Fermi surface, the attenuation decreases with temperature as a power law reflecting the spectrum of low-energy excitations near the nodes of the order parameter.<sup>48,49,56,57</sup> Impurity scattering modifies this spectrum near the nodes, and at low energies a new energy scale,  $\gamma \ll \Delta_0$ , appears, which is roughly the 'bandwidth' of low-energy quasiparticles bound to the impurity distribution by Andreev scattering.<sup>41</sup> The bandwidth also appears as an impurity-renormalized quasiparticle width at zero energy,  $\tilde{\epsilon}^R(0) = i\gamma$ . This new low energy scale defines a cross-over from the power law behavior associated with scattering of continuum quasiparticles for  $\gamma/k_B < T \ll T_c$ , to a temperature independent attenuation in the limit  $k_B T \ll \gamma$ .<sup>48,49,56,57</sup>

The bandwidth of the impurity-induced Andreev levels is determined by the self-consistency equation for the quasiparticle self-energy,

$$\gamma = \Gamma_u \frac{\langle \gamma [|\Delta(\mathbf{p}_f)|^2 + \gamma^2]^{-\frac{1}{2}} \rangle}{\cot^2 \delta_0 + \langle \gamma [|\Delta(\mathbf{p}_f)|^2 + \gamma^2]^{-\frac{1}{2}} \rangle^2}, \quad (7)$$

where  $\langle \dots \rangle$  is an average over the Fermi surface and  $\Gamma_u = n_{\text{imp}}/\pi N_f$  is the scattering rate in the normal state for resonant impurities and  $\delta_0$  is the scattering phase shift. For the high purity UPt<sub>3</sub> crystals studied in Ref. 10, i.e., low scattering rate, the crossover temperature is very low compared to  $T_c$ ; analysis of the thermal conductivity provides a determination of both the scattering phase shift as well as the bandwidth of the impurity-induced Andreev states. The scattering centers are nearly resonant, i.e.,  $\delta_0 \simeq \pi/2$ , giving a bandwidth, and cross-over temperature, of order  $\gamma \sim k_B T^* \approx 0.2\sqrt{\mu\Gamma_u\Delta_0} \approx 0.07k_B T_c$ , where  $\Gamma_u \simeq 0.03k_B T_c$ ,  $\Delta_0 \simeq 2.0k_B T_c$  and the slope of the excitation gap near the line node is  $\mu = \Delta_0^{-1} |d\Delta/d\vartheta|_{\vartheta=\pi/2} \simeq 2$ . Thus, transport experiments on UPt<sub>3</sub> have so far not investigated the ultra-low temperature region  $k_B T \ll \gamma$  in any systematic way.

## V. TRANSVERSE SOUND ATTENUATION

The anisotropy and temperature dependence of the sound attenuation is sensitive to the polarization of the sound field and the symmetry of the order parameter. This was the basis of transverse sound attenuation experiments that provided early evidence for a line of nodal excitations in the basal plane.<sup>47</sup> We examine the *ab*-plane anisotropy of the transverse sound attenuation. Our analysis covers the full temperature range below  $T_c$ , and is particularly sensitive to the polarization and anisotropy of the order parameter for both A and B phases of UPt<sub>3</sub>. To illustrate the sensitivity of the transverse sound polarization to the order parameter symmetry consider the theoretical models for the A phase of UPt<sub>3</sub>. The *ab*-plane anisotropies of the excitation gap,  $|\Delta(\vartheta \neq \pi/2, \phi)|$ , for the A phase of four pairing models, E<sub>2u</sub>, E<sub>1g</sub>, A<sub>2u</sub>  $\oplus$  B<sub>1u</sub> and A<sub>1g</sub>  $\oplus$  E<sub>1g</sub>, are shown in Fig. 2.

The propagation and polarization vectors determine the angular dependence of the momentum transport by quasiparticles on the Fermi surface; the matrix element is proportional to  $[\mathbf{p}_f]_i^2 [\mathbf{v}_f]_j^2$ . This angular dependence is weighted by the angle-resolved density of states for momentum transfer via impurity scattering, which depends on the anisotropy of the quasiparticle excitation spectrum through  $|\Delta(\mathbf{p}_f)|^2$ . When both the propagation and polarization vectors are in the basal plane ( $\mathbf{q} \parallel \mathbf{a}, \mathbf{e} \parallel \mathbf{b}$ )<sup>58</sup> the matrix element is proportional to  $\sin^2 2\phi$ , and is maximum at angles of  $\pi/4$  from these two axes, i.e., the midpoints between the polarization and propagation directions. If these midpoint directions coincide with nodal directions (e.g.  $|\Delta(\mathbf{p}_f)| \sim |\cos 2\phi|$  for E<sub>2u</sub>) then the attenuation will be a maximum, while if the midpoint directions are along the antinodal directions then the attenuation is a minimum. This is illustrated in Fig. 2 where the polarization is directed along the  $\mathbf{a}$  direction. The attenuation is largest when the polarization is along an antinode of the order parameter, and it is smallest

when the polarization is along a nodal direction. One can immediately see that we should expect to observe a rather different *ab*-plane angular dependence to the attenuation for the different order parameter models proposed for UPt<sub>3</sub>. We quantify these remarks below.

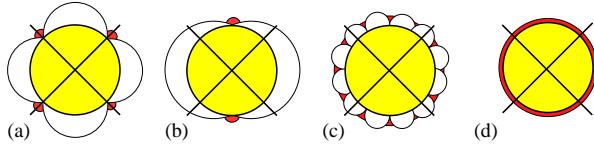


FIG. 2. Sketch of the  $\phi$  dependence of the A phase pairing states at  $\vartheta \neq \pi/2$  for (a)  $E_{2u}$ , (b)  $E_{1g}$ , (c)  $A_{2u} \oplus B_{1u}$ , and (d)  $A_{1g} \oplus E_{1g}$  models. The crossed lines represent the areas of maximal absorption of a sound probe with  $\mathbf{ab}$  symmetry ( $\mathbf{q} \parallel \mathbf{a}, \mathbf{\epsilon} \parallel \mathbf{b}$ ) at the Fermi surface. The dark shaded areas represent the distribution of quasiparticle excitations.

### A. Results

In order to make quantitative predictions for UPt<sub>3</sub> we use heat capacity and thermal conductivity measurements to fix the magnitude of the order parameter, the Fermi surface anisotropy, the nodal parameters and the scattering rate, all of which control the temperature dependence and anisotropy of the thermal conductivity below  $T_c$ .<sup>25,40</sup> We used variational basis functions based on the polynomial functions in Table I. Symmetry dictates the geometry of the line and point nodes, as well as the topological indices for the point nodes, but not the slopes or curvature of the nodal regions of the excitation gap. Our variational procedure fits these parameters to the low-energy excitation spectrum from the thermal conductivity data of Ref. 10. The order parameter is then determined self-consistently. For a detailed discussion of this analysis see Refs. 25,40. The anisotropy and temperature dependence of the transverse sound attenuation are then calculated with no additional parameters or adjustments of the order parameter models shown in Fig. 2.

The transition temperature and splitting of the zero-field transition in UPt<sub>3</sub> determine the scale and relative magnitudes of the two order parameter components; they determine the dominant and subdominant instability temperatures. The instability temperature for the dominant channel is the transition temperature,  $T_{c1} = T_{c+}$ , while the second instability temperature represents the strength of the subdominant pairing channel,  $T_{c2} \propto \omega_c \exp(-1/V_2)$ , where  $V_2$  is the pairing interaction for the sub-dominant channel and  $\omega_c$  is the cut-off energy.<sup>59</sup> The physical transition temperature,  $T_{c-}$ , separating the A and B phases depends on the relative strength of the two paring channels, i.e., on  $T_{c2}/T_{c+} \leq 1$ . We solve the gap equation for the second transition and adjust  $T_{c2}$  to the observed splitting in the specific heat,  $(T_{c+} - T_{c-})/T_{c+} \approx 60 \text{ mK}/495 \text{ mK}$ .<sup>12,9</sup> All other mate-

rial parameters are taken from our previous analysis of the thermal conductivity of UPt<sub>3</sub>.<sup>40</sup> We show the quality of the theoretical fits to the thermal conductivity,  $\kappa$ , and heat capacity for the  $E_{2u}$  model in Fig. 3. Similar fits can be obtained for the  $E_{1g}$  and AE models. However, only the in-plane thermal conductivity data can be accounted for by the AB model.

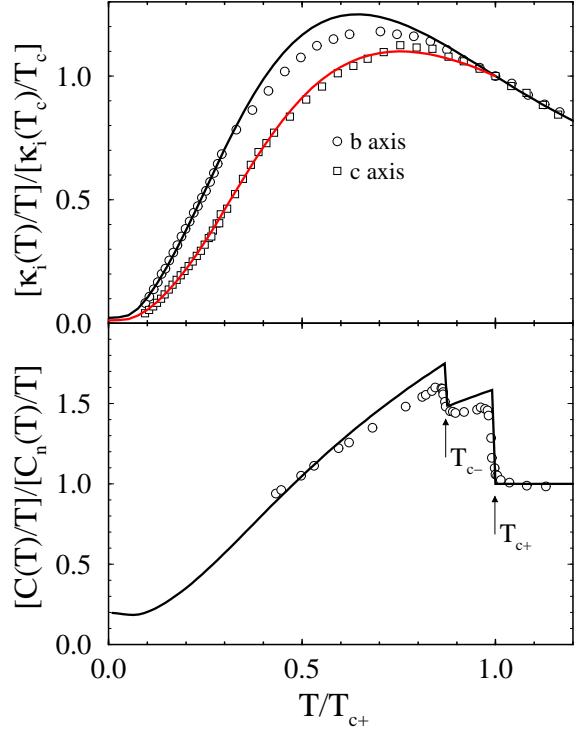


FIG. 3. Fit of the  $E_{2u}$  model to the thermal conductivity data (top)<sup>10</sup> and the specific heat data (bottom).<sup>9</sup>

#### 1. Identification of the A-phase

In Fig. 4 we show the attenuation data reported by Ellman *et al.*<sup>12</sup> for transverse sound propagation in the *ab*-plane with  $\mathbf{q} \parallel \mathbf{a}$  and polarizations both in- and out-of-plane,  $\mathbf{\epsilon} \parallel \mathbf{b}$  and  $\mathbf{\epsilon} \parallel \mathbf{c}$ . These measurements were made on the same batch of crystals of UPt<sub>3</sub> as the heat capacity and thermal conductivity measurements shown in Fig. 2. In addition to the polarization, the data show a pronounced change in the anisotropy and temperature dependence at the A-B phase transition.

The enhanced absorption in the A phase compared to that of the B phase at the same temperature results from excess quasiparticles that scatter off the impurity distribution due to additional nodes of the A phase order parameter compared to that of the B phase. Below  $T_{c-}$  the subdominant order parameter closes the additional nodes of the A phase order parameter. Thus, the sound absorption drops faster in the B phase than it would in the A phase. The experimental results for the sound attenuation, including the anomaly at  $T_{c-}$ , are in excellent agree-

ment with theoretical calculations for the  $E_{2u}$  model of the A and B phases, with an A phase given by a  $\eta = (1, 0)$  state, but not a  $(0, 1)$  state; and a B phase at low temperature which is (approximately) the  $\eta = (1, \pm i)$  state.<sup>60</sup> The comparison between theory and experiment is unsatisfactory for all other models. Even including the frequency dependence of the viscosity,  $\eta_{ij,ij}(\mathbf{q}, \omega)$ , does not change this result. We find only minor corrections to the sound attenuation for  $\omega \ll \Delta_0$  (see Fig. 4). The  $(1, 0)$  state of the A phase, as determined by sound attenuation, also agrees with the order parameter orientation obtained from the observed six-fold oscillations of  $H_{c2}(\phi)$ , and the change in sign of these oscillations when crossing the A-C phase boundary.<sup>61</sup>

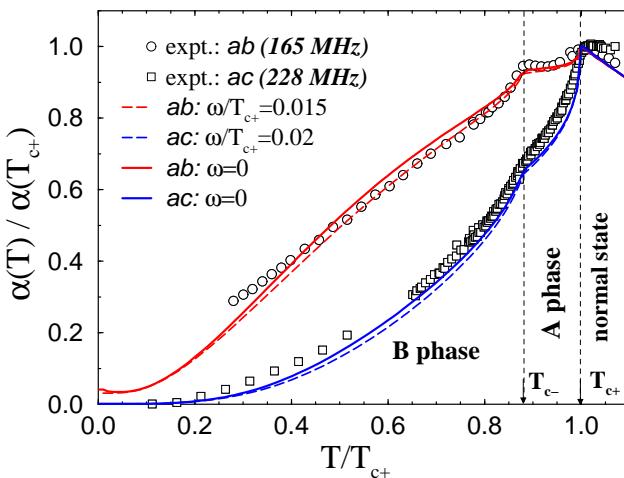


FIG. 4. Comparison between the measured and calculated transverse sound attenuation of  $\text{UPt}_3$ . The theoretical calculation is for the  $E_{2u}$  model and the phenomenological model previously used for the total elastic and inelastic scattering rate,  $\Gamma(T) = 0.03k_B T_{c+}(1 + T^2/T_{c+}^2)$ , obtained from the thermal conductivity analysis. The data are from Ellman *et al.*,<sup>12</sup> which are corrected to vanish at  $T = 0$ .

In Fig. 5 we show the sound attenuation for **ab** and **ac** polarizations and different pairing models. Note in particular that none of the order parameter models shown in Fig. 5 can account for both anomalies in  $\alpha_{\text{ab}}$  and  $\alpha_{\text{ac}}$  at  $T_{c-}$ . The main result of this work is that the experimental data for the heat capacity, thermal conductivity, and sound attenuation, as well as the H-T phase diagram, are explained *only* by an order parameter with an orbital  $E_{2u}$  pairing symmetry. We emphasize that there are no adjustable parameters in the calculation of the sound attenuation; all parameters of the model were previously determined by fitting the theoretical model parameters to the heat capacity and thermal conductivity.<sup>40</sup>

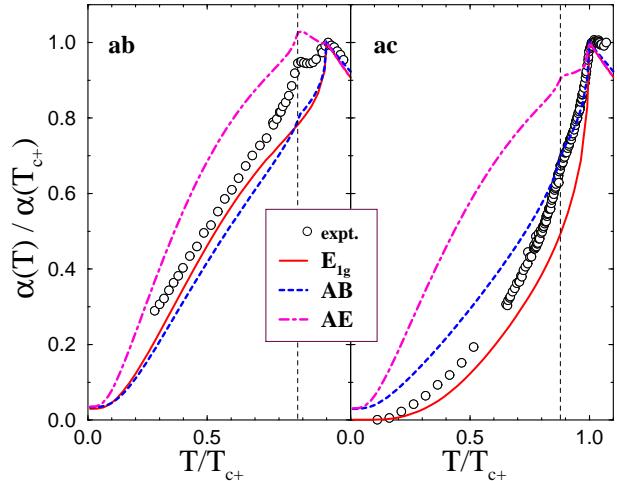


FIG. 5. Attenuation of **ab** and **ac** transverse sound for the various order parameter models.

## 2. Domain Structure

Neutron diffraction studies in pure  $\text{UPt}_3$  under pressure,<sup>20</sup> and at ambient pressure in Pd doped samples ( $\text{Pt} \leftrightarrow \text{Pd}$ ),<sup>62</sup> demonstrated that the splitting of  $T_c$  correlates with the basal plane AFM, suggesting a SBF coupling between the superconducting and AFM order. It has been argued, based on neutron diffraction studies as a function of magnetic field, that the AFM order is described either by a distribution of three equally populated domains, with  $\mathbf{Q}$  vectors oriented  $120^\circ$  to one another, or a triple- $\mathbf{Q}$  structure.<sup>63,64</sup> If a domain structure is present then, in the AFM model for the SBF, the superconducting order parameter describing the A phase in the E-rep models may also form a domain structure. Such a domain structure leads to a weakening of the anomaly for  $\alpha_{\text{ab}}$  at  $T_{c-}$ , but leaves the anomaly in  $\alpha_{\text{ac}}(T_{c-})$  virtually unchanged.

In Fig. 6 we show the effect of multi-domain averaging on the anisotropy of the transverse sound attenuation. These calculations show that an AFM domain structure does not destroy the anomaly in the attenuation at  $T_{c-}$ . However, averaging over domains suppresses the characteristic enhancement of  $\alpha_{\text{ab}}$  for the  $E_{2u}$  order parameter coupled to a dominant or single domain of the SBF. In particular, the domain-averaged attenuation,  $\langle \alpha_{\text{ab}} \rangle$ , for the  $E_{2u}$  pairing state drops roughly twice as fast as the measured attenuation in the A-phase. If the AFM order parameter is the SBF for the superconducting phases, then our calculations are in agreement with transport and heat capacity measurements only for an  $E_{2u}$  order parameter coupled to a dominant domain, or a triple- $\mathbf{Q}$  structure for the antiferromagnetic SBF.<sup>24</sup>

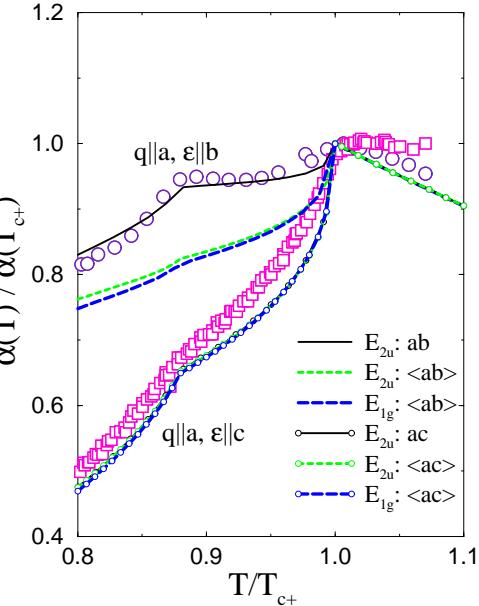


FIG. 6. Configuration averaged attenuation assuming three equally populated mono-domains for an  $E_{1g}$  and  $E_{2u}$  pairing state. For a comparison the results for the  $E_{2u}$  state of Fig. 5 are re-plotted.

### 3. Scattering phase shifts

It has been pointed out in several studies that the temperature dependence of the transport coefficients in  $\text{UPt}_3$  is qualitatively consistent with strong scattering in the unitarity limit. Our analysis of the thermal conductivity data,<sup>10</sup> and the attenuation data<sup>12</sup> confirm that the scattering phase shift is near the resonant limit; from the analysis of the thermal conductivity we obtain  $\delta_0 \geq 80^\circ$ ,<sup>25</sup> while the transverse sound attenuation data implies a scattering phase shift  $\delta_0 > 60^\circ$ , as shown in Fig. 7.

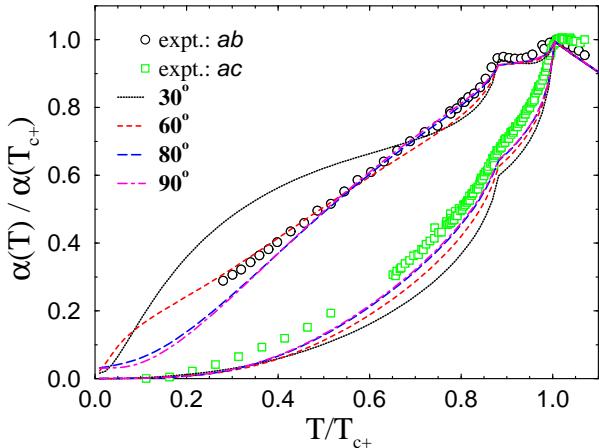


FIG. 7. Sensitivity of the transverse sound attenuation to the scattering phase shift  $\delta_0 = 30^\circ, 60^\circ, 80^\circ, 90^\circ$  for the  $E_{2u}$  pairing model and the same parameters as shown in Fig. 4.

### 4. Ultra-low temperature region

The temperature dependence of the transverse sound attenuation is predicted to change qualitatively below the cross-over temperature,  $k_B T < \gamma$ . The zero-temperature limit is finite, reflecting the finite density of states at the Fermi level, and the leading temperature-dependent corrections are of the Sommerfeld type,  $\mathcal{O}[(T/\gamma)^2]$  for  $k_B T < \gamma$ . The limiting attenuation in the ultra-low temperature limit,  $k_B T \ll \gamma$ , is obtained from the viscous stress tensor for  $T \rightarrow 0$  and  $\omega \rightarrow 0$ . In this limit the states contributing to the stress tensor in Eq. (5) are confined to the impurity-induced Andreev band of order  $\gamma \gg \max(T, \omega)$ . The spectrum and self-energy are weakly energy dependent on this scale, thus, we can evaluate the slowly varying parts of the integrand in Eq. (6) at zero energy to obtain

$$\eta_{ij,ij} \simeq N_f \int d\mathbf{p}_f [\mathbf{v}_f]_i^2 [\mathbf{p}_f]_j^2 \frac{\gamma^2}{[(\Delta(\mathbf{p}_f))^2 + \gamma^2]^{\frac{3}{2}}}, \quad (8)$$

where we have neglected the Sommerfeld corrections of order  $\mathcal{O}[(T/\gamma)^2]$ .<sup>65</sup>

Whether or not the zero-temperature limit is *universal*, i.e., independent of the density and scattering cross-section for the impurities, depends on the polarization of the acoustic wave and the symmetry of the ground state. The damping is universal only for transverse waves propagating along the high symmetry directions in the  $ab$ -plane, i.e., for polarization  $\epsilon \parallel \mathbf{b}$  propagating along  $\mathbf{q} \parallel \mathbf{a}$ , or vice versa. For these polarizations the ground state of *all* the models we discuss, except the AE-model, possess a *universal* limit for the attenuation. Table II summarizes the results for the  $E_{1g}$  and  $E_{2u}$  models. For the ground state of either E-representation we find  $\eta_{\mathbf{ab},\mathbf{ab}} \simeq v_f^2 p_f^2 N_f / (8\mu\Delta_0)$ , where  $\mu \approx 2$  is the slope of the excitation gap near the line node in the  $ab$ -plane. For any other polarization the relevant viscosity is non-universal for  $\omega, T \ll \gamma$ . For example, for the  $E_{2u}$  ground state we obtain a limiting value for  $\alpha_{\mathbf{ac}}(0) \approx \frac{2\mu\gamma}{\mu_2^2\Delta_0} \alpha_{\mathbf{ab}}(0) \ll \alpha_{\mathbf{ab}}(T_{c+})$ , where  $\mu_2 \approx 4$  is the parameter defining the curvature of the excitation gap near the quadratic point node along the  $\mathbf{c}$  axis.<sup>66</sup>

Further experiments using transverse sound can be used to confirm the predictions for the  $ab$ -plane anisotropy of the order parameter in  $\text{UPt}_3$ . The ideal method would be to propagate transverse sound along the  $\mathbf{c}$  axis and measure the attenuation as a function of the azimuthal orientation of the polarization in the  $ab$ -plane. The qualitative predictions for the in-plane anisotropy can be deduced from Fig. 2; a two-fold symmetry of the anomaly at  $T_{c-}$  is expected for the  $E_{1g}$  representation and a nearly isotropic attenuation above  $T_{c-}$  for the other models. In the B phase all models show a nearly isotropic attenuation except for the AE model,

which has a two-fold symmetry. A similar experiment was suggested by Moreno and Coleman<sup>67</sup> to map out the gap structure in the high- $T_c$  cuprates.

TABLE II. Asymptotic low-temperature limits of the sound attenuation ( $\omega, T \rightarrow 0$ ) and thermal conductivity for the  $E_{1g}$  and  $E_{2u}$  pairing states. The results are scaled in units of  $\kappa_0 = (\pi^2/3)k_B^2 T N_f v_f^2 \tau_\Delta$  and  $\alpha_0 = (\omega^2/4\varrho c_s^3) p_f^2 N_f v_f^2 \tau_\Delta$ , where  $\tau_\Delta \equiv \hbar/2\mu\Delta_0(0)$  is an effective transport time.

Transport Coefficient	$E_{1g}$	$E_{2u}$
$\kappa_{bb}/\kappa_0$	1	1
$\kappa_{cc}/\kappa_0$	$2\mu\gamma/(\mu_1^2\Delta_0)$	$\mu/\mu_2$
$\alpha_{ab}/\alpha_0$	1	1
$\alpha_{ac}/\alpha_0$	$\frac{8\mu}{1+2\mu^2} \frac{\Gamma_u}{\Delta_0}^*$	$\frac{2\mu}{\mu_2^2} \frac{\gamma}{\Delta_0}$

\* In the strong scattering limit including vertex corrections.

To summarize the main conclusions, we have shown that transverse ultrasound provides detailed information on the orbital pairing symmetry of both the superconducting A and B phases of UPt<sub>3</sub>; the anisotropy and the anomalies in the temperature dependence of the attenuation for different polarizations is explained *only* by an  $E_{2u}$  order parameter. Measurements of  $\alpha_{ab}$  and  $\alpha_{ac}$  are in excellent agreement with a (1, 0) state for the A phase, corresponding to a  $p_z(p_x^2 - p_y^2)$  order parameter. Further measurements at lower temperatures, or as a function of impurity disorder, may also be used to test the prediction of a universal limit for the in-plane transverse sound attenuation.

## VI. ACKNOWLEDGMENTS

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## APPENDIX A:

In this appendix we summarize the relevant nonequilibrium transport equations for our calculations of the sound attenuation in unconventional superconductors. For a more extensive review of nonequilibrium transport theory in superconductors see Refs. 41,68.

The nonequilibrium (Keldysh) Green's function,

$$\delta\hat{g}^K = \delta\hat{g}^R \circ \Phi_0 - \Phi_0 \circ \delta\hat{g}^A + \delta\hat{g}^a, \quad (A1)$$

contains the spectral response given by the retarded and advanced Green's functions ( $\delta\hat{g}^{R,A}$ ), and the anomalous

response given by  $\delta\hat{g}^a$ , which in normal metals is essentially the nonequilibrium distribution function. The equilibrium distribution function is  $\Phi_0 = \tanh(\epsilon/2k_B T)$ , and we use the shorthand notation for the shifting product,  $\Phi_0 \circ A = \Phi_0(\epsilon - \omega/2)A(\epsilon, \omega)$  and  $A \circ \Phi_0 = A(\epsilon, \omega)\Phi_0(\epsilon + \omega/2)$ . Pairing correlations and particle-hole coherence require a matrix structure for the particle-hole degree of freedom. The 'hat' over the Green's functions and self-energies indicates their  $4 \times 4$  matrix structure in particle-hole and spin space.

The transport equations for the Green's functions, linearized with respect to an external perturbation, are

$$[\delta\hat{g}^{R,A}, \hat{h}^{R,A}]_o = [\hat{g}_0^{R,A}, \hat{\sigma}_{\text{ext}} + \delta\hat{\sigma}^{R,A}]_o, \quad (A2)$$

$$\begin{aligned} \hat{h}^R \circ \delta\hat{g}^a - \delta\hat{g}^a \circ \hat{h}^A &= \delta\hat{\sigma}^a \circ \hat{g}_0^A - \hat{g}_0^R \circ \delta\hat{\sigma}^a \\ &- [\hat{\sigma}_{\text{ext}}, \Phi_0]_o \circ \hat{g}_0^A - \hat{g}_0^R \circ [\Phi_0, \hat{\sigma}_{\text{ext}}]_o, \end{aligned} \quad (A3)$$

where  $\hat{h}^{R,A} = \epsilon\hat{\tau}_3 - \hat{\sigma}_0^{R,A}$ ,  $\hat{\tau}_3$  is a Pauli matrix, and  $\hat{\sigma}_{\text{ext}}$  is the external perturbation, e.g., the coupling of quasiparticles to a sound field (ionic displacement field). The transport equations determine the deviations of the Green's functions from their local equilibrium values,  $\delta\hat{g}^X = \hat{g}^X - \hat{g}_0^X$ , in terms of the external field and the corrections to the self-energies,  $\delta\hat{\sigma}^X = \hat{\sigma}^X - \hat{\sigma}_0^X$  with  $X \in \{R, A, K\}$ . The anomalous self-energy,  $\delta\hat{\sigma}^a$ , is defined similarly to the  $\delta\hat{g}^a$ ,

$$\delta\hat{\sigma}^K = \delta\hat{\sigma}^R \circ \Phi_0 - \Phi_0 \circ \delta\hat{\sigma}^A + \delta\hat{\sigma}^a. \quad (A4)$$

The equilibrium Green's functions,  $\hat{g}_0^X$ , and self-energies,  $\hat{\sigma}_0^X$ , are inputs to the linearized transport equations. At low temperatures, and for long-wavelength, low-frequency sound, the damping of the acoustic wave is determined by the scattering of quasiparticles off impurities and defects that are co-moving with the ionic lattice.<sup>53</sup> Impurity scattering enters the transport equations via the impurity-scattering self energies,

$$\hat{\sigma}^X(\mathbf{p}_f; \epsilon, t) = n_{\text{imp}} \hat{t}^X(\mathbf{p}_f, \mathbf{p}_f; \epsilon, t), \quad (A5)$$

where the quasiparticle-impurity scattering t-matrices are given by

$$\begin{aligned} \hat{t}^{R,A}(\mathbf{p}_f, \mathbf{p}'_f; \epsilon, t) &= \hat{u}(\mathbf{p}_f, \mathbf{p}'_f) + N_f \langle \hat{u}(\mathbf{p}_f, \mathbf{p}''_f) \circ \\ &\hat{g}^{R,A}(\mathbf{p}''_f; \epsilon, t) \circ \hat{t}^{R,A}(\mathbf{p}''_f, \mathbf{p}'_f; \epsilon, t) \rangle_{\mathbf{p}''_f}, \end{aligned} \quad (A6)$$

$$\begin{aligned} \hat{t}^K(\mathbf{p}_f, \mathbf{p}'_f; \epsilon, t) &= N_f \langle \hat{t}^R(\mathbf{p}_f, \mathbf{p}''_f; \epsilon, t) \circ \\ &\hat{g}^K(\mathbf{p}''_f; \epsilon, t) \circ \hat{t}^A(\mathbf{p}''_f, \mathbf{p}'_f; \epsilon, t) \rangle_{\mathbf{p}''_f}. \end{aligned} \quad (A7)$$

The t-matrices are calculated self-consistently with the order parameter,  $\hat{\Delta}(\mathbf{p}_f)$ , and with the impurity vertex,  $\hat{u}(\mathbf{p}_f, \mathbf{p}'_f)$ , describing elastic coupling of quasiparticles to impurities. The calculations presented here assume isotropic, non-magnetic impurities with  $\hat{u} = u_0 \hat{1}$  for all  $(\mathbf{p}_f, \mathbf{p}'_f)$ . This model is then described by two

parameters, e.g., the s-wave scattering phase shift,  $\delta_0 = \tan^{-1}(\pi N_f u_0)$ , and the density of impurities,  $n_{\text{imp}}$ . Another useful parametrization is in terms of the cross-section,  $\sigma = (4\pi/k_f^2) \sin^2 \delta_0$ , and the normal-state scattering rate in the unitarity limit ( $\delta_0 = \pi/2$ ),  $\Gamma_u = n_{\text{imp}}/\pi N_f$ . Note that the transport scattering rate in this model is given by  $1/2\tau_0 = \Gamma_u \sin^2 \delta_0$ , and the elastic mean free path is then  $\ell_{\text{el}} = v_f \tau_0$ .

The other key term entering the transport equations is the off-diagonal pairing self energy, or order parameter. The general form for the pairing self energies is

$$\hat{\Delta}^{R,A} = \begin{pmatrix} 0 & \Delta i\sigma_y + \Delta \cdot i\sigma\sigma_y \\ \bar{\Delta} i\sigma_y + \bar{\Delta} \cdot i\sigma_y \sigma & 0 \end{pmatrix}, \quad (\text{A8})$$

where the spin-singlet ( $\Delta$ ) and spin-triplet ( $\Delta$ ) order parameters are given by the gap equations,

$$\Delta^{R,A}(\mathbf{p}_f; t) = \int \frac{d\epsilon}{4\pi i} \langle V^s(\mathbf{p}_f, \mathbf{p}'_f) f^K(\mathbf{p}'_f; \epsilon, t) \rangle_{\mathbf{p}'_f}, \quad (\text{A9})$$

$$\Delta^{R,A}(\mathbf{p}_f; t) = \int \frac{d\epsilon}{4\pi i} \langle \underline{\mathbf{V}}^t(\mathbf{p}_f, \mathbf{p}'_f) \cdot \mathbf{f}^K(\mathbf{p}'_f; t) \rangle_{\mathbf{p}'_f}, \quad (\text{A10})$$

where  $V^s$  and  $\underline{\mathbf{V}}^t$  are the pairing interactions in the singlet and triplet channels. The components  $\bar{\Delta}$  and  $\Delta$  are related to  $\Delta$  and  $\Delta$  by fundamental symmetries (see Appendix C of Ref. 69); in equilibrium  $\bar{\Delta} = \Delta^*$  and  $\bar{\Delta} = \Delta^*$ .

To complete the set of equations for the linear response equations we write the solutions for the equilibrium response functions in terms of renormalized quasiparticle energy and order parameter. For spin-singlet and unitary spin-triplet pairing the general solutions for the equilibrium retarded and advanced Green's functions are

$$\hat{g}_0^{R,A} = -\pi \frac{\tilde{\epsilon}^{R,A} \hat{\tau}_3 - \hat{\Delta}^{R,A}}{\sqrt{|\tilde{\Delta}^{R,A}|^2 - (\tilde{\epsilon}^{R,A})^2}}, \quad (\text{A11})$$

where the renormalized quasiparticle energy is

$$\tilde{\epsilon}^{R,A}(\mathbf{p}_f, \epsilon) = \epsilon - \frac{1}{4} \text{Tr} [\hat{\tau}_3 \hat{g}_{\text{imp}}^{R,A}(\mathbf{p}_f, \epsilon)]. \quad (\text{A12})$$

For resonant s-wave scattering

$$\hat{g}_{\text{imp}}^{R,A}(\epsilon) = -\Gamma_u \left\langle \hat{g}_0^{R,A}(\mathbf{p}_f; \epsilon) \right\rangle_{\mathbf{p}_f}^{-1}. \quad (\text{A13})$$

For s-wave scattering the renormalization of the off-diagonal self-energy by impurity scattering vanishes,

$$\hat{\Delta}^{R,A}(\mathbf{p}_f, \epsilon) = \hat{\Delta}(\mathbf{p}_f), \quad (\text{A14})$$

for non-identity representations of the point group, i.e., excluding the  $A_{1g}$  representation. Finally, we note that the diagonal component of the nonequilibrium Green's function that determines the momentum stress tensor in Eq. (5) is obtained from the matrix Green's function by  $\delta g^K = \frac{1}{4} \text{Tr} \delta \hat{g}^K$ .

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